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Development of a multimedia model (IIAQ-CC) to assess climate change influences on volatile and semi-volatile organic compounds in indoor environments

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ABSTRACT

An indoor air quality model (IIAQ-CC) was developed to assess climate change (CC) influences on indoor level of volatile or semi-volatile organic compounds. With IIAQ-CC, the CC influences is predicted in integrated manners by factors including fate/transport properties of pollutants, building characteristics, occupant behaviors, indoor emission sources, and outdoor air quality. IIAQ-CC was evaluated for the temperature sensitivity of indoor formaldehyde (FA) concentration at outdoor temperature below 20 °C. No significant difference (*t*-test, $p < 0.05$) was found between the predicted (3.9 ± 1.0%/°C) and the measured (5.0%/°C with a 95% confidence interval (3.7%/°C to 6.4%/°C)), demonstrating that IIAQ-CC can successfully predict CC influences driven by temperature change on VOCs of indoor environmental concern. IIAQ-CC predicts that the long-term FA concentration may slowly rise due to increase in the outdoor concentration or may decrease in the presence of strong indoor sources in RCP8.5 conditions of South Korea. In either case, the long-term change does not exceed 12% of the average FA concentration of the base period. Over a shorter period (e.g., a month), the FA level may noticeably rise (by about 4 times) when windows are closed due to exceedingly hot or cold weather. The FA concentration change is primarily governed by duration of windows kept open, which depends on outdoor temperature. That is, temperature manifests its effects on the indoor air quality essentially by change in window opening behavior rather than by changes in kinetics or equilibrium processes such as photo-degradation and sorption/desorption onto/from walls and floor.

1. Introduction

Climate change (CC) is likely to cause increase in the average earth surface temperature, incidence of heat and cold waves, and extreme precipitation throughout the 21st century [1]. These changes have the potential to alter not only the quality of atmosphere but that of indoor air [2]. Since people spend 80%–90% of their time indoors, it is important to assess CC influences on the exposure to indoor air pollutants.

The factors influencing indoor air pollutant levels in response to CC can be classified into three categories; (1) fate and transport properties of pollutants, (2) building characteristics such as size, number of window, cracks, interior surface materials, availability of heater/cooler system, and indoor air cleaner, etc., and (3) occupant behavior [3].

The first category refers to the fact that the level of pollutants in indoor air will depend on pollutants' fate and transport properties. The same is true of ambient air, which also influences the indoor air quality via indoor-outdoor air exchange. For example, the monthly average concentration of the PAHs in the atmosphere was predicted to increase up to a factor of $\sim 10^2$ in South Korea during the period of 2000–2049 under the A1B scenario [4]. However, the degree to which the concentration changes varied among the PAHs depending on their fate and transport properties.

The second category regards the building factors including use of mechanical equipment such as air-conditioner and/or indoor air cleaner, the number and size of windows, and type of interior surface materials. In particular, use of the heating, ventilating, and air conditioning (HVAC) systems may significantly alter the level of indoor air pollution by changing the ventilation rate. Likewise, natural ventilation influences the indoor air pollution. Ilacqua et al. [5] predicted that the air exchange through crack would decrease by 5% in the period of 2040–2070 from that of 1970–2000 under the A1 scenario. The decrease in the air exchange rate will reduce the indoor-outdoor exchange

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flux of gaseous pollutants, which contributes to increasing (or decreasing) the gaseous pollutants level indoor if the indoor level is originally higher (or lower) than that of outdoor. Lee et al. [6] estimated that the particulate matters of aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$) entering indoor through cracks and openings would increase by 7% in naturally ventilated homes with the ambient air temperature rise of 2 °C–3 °C. The increased $PM_{2.5}$ infiltration implies more infiltration of pollutants sorbed on the particulate matters (PMs) (e.g., hydrophobic semi-volatile organic compounds (SVOCs)). However, the mass of a sorbed pollutant per unit mass of PMs would mostly decrease with temperature rise, indicating that the temperature effect on the infiltration rate of sorbed pollutants is not straightforward. Besides, the rate of emission of volatile organic compounds (VOCs) and SVOCs from interior surfaces or their sorption equilibrium between air and the indoor surfaces are strongly influenced by temperature [7–10].

The third category concerns the adjustment or alteration in occupant's behavior to adapt to CC. In particular, window opening behavior coupled with the use of HVAC system as influenced by weather change will have a certain impact on the indoor air quality through the change in ventilation rate [11] and indoor temperature.

All the factors in the three categories will interact with one another in complex manners to affect the indoor pollutant levels. To predict the CC influences and resulting change in indoor air quality, therefore, these factors are required to be simultaneously taken into account in quantitative and integrated manners. However, existing studies have been somewhat limited in meeting the requirement. For instance, describing the effects of CC on indoor air quality have been non-specific and/or qualitative in many of existing studies [2,3,12–14]. It is only recent that a few quantitative studies [5,6] appeared. However, these studies focused on crack infiltration or natural ventilation to provide in depth understanding of potential CC influences only on the specific processes of concern. Some studies predicted the changes in air quality of SVOCs and VOCs as a result of CC in outdoor [4,15–18]. While these studies are valuable as outdoor air quality influences indoor air quality, attempt has yet to be made to link their results to indoor air quality. Several studies modeled both vapor and PMs phases indoors [7,8,19–21], their results were not extended to potential effects of temperature change induced by CC on the vapor-PMs partitioning. Besides, numerous studies investigated individual factors including building characteristics (air change rate and use of HVAC system) [22,23], occupant behavior (window opening/closing) [24,25], and sources (indoor and outdoor) [19,26], but not necessarily with regard to CC influences. To our knowledge, however, in no study were simultaneously integrated all these factors to assess the CC influences on the indoor air quality.

Recognizing these limitations of existing studies, the main objective of the present work was to develop an indoor air quality model (Integrated Indoor Air Quality Model under Climate Change, IIAQ-CC) which is capable of quantitatively predicting the influence of CC on indoor air quality. Specifically, IIAQ-CC was built and its performance was assessed against the measured data of formaldehyde (FA). Subsequently, a case study was presented where the influence of CC was predicted by using IIAQ-CC on the change in the level of FA in indoor air under the RCP8.5 scenario.

2. Methods

2.1. IIAQ-CC

IIAQ-CC is a dynamic multimedia model that predicts the level of a pollutant in indoor media of a well-mixed space by taking into account fate and transport processes for an indoor air compartment and an interior surface compartment (Fig. 1). The indoor air compartment consists of two sub-compartments (vapor and PMs) which are assumed to be in chemical equilibrium with each other. The interior surface compartment can include various sub-compartments such as the surfaces of floor, wall, ceiling, and furniture. These individual surface sub-compartments are divided into a number of fine sub-layers (10 for the floor in the present work) in each of which a uniform pollutant concentration is assumed for numerical analysis. IIAQ-CC was programmed with Visual Fortran (v. 2011) using the Microsoft visual studio (v. 2010) on the Windows operating system.

2.1.1. Fate and transport processes

The main processes include mechanical and natural ventilations (through window and crack), pollutant partitioning between vapor and solid phases (PMs and the interior surface compartment), photochemical degradation of vapor phase pollutants, deposition/resuspension of PMs (and associated pollutants) to/from the surface compartment, and removal of PMs (and associated pollutants) with air handling system (AHS) and recirculation (RCL) filters and of PMs on the surfaces by clean up. The process equations used in IIAQ-CC are summarized in Table A.1 of the supplementary data (SD). By adopting the process equations, a mass balance equation was formed for each of all the sub-compartments. For example, equations (1) and (2) below are typical mass balance equations used in IIAQ-CC for a pollutant in the vapor and PMs phases in the air compartment, respectively. The mass balance equations can be readily modified as necessary.

$$V_c\cdot\frac{d[C_V]}{dt} = (Q_{mv}\cdot C_{V_o} + Q_{ra}\cdot C_V)\cdot(1-e) + (Q_{cf} + Q_{window})\cdot C_{V_o} + Q_{ADJ_o}\cdot C_{V_{ADJ_o}} - [(Q_{mv} + Q_{ra} + Q_{cf} + Q_{ADJ_o} + Q_{window})\cdot C_V] - (\sum_i E_{S_i}\cdot A_i + V_c\cdot k_v\cdot C_V + Q_{rcl}\cdot e_{rcl}\cdot C_V) + \sum_j E_{V_j} \quad (1)$$

where V_C is the volume of air compartment (m^3); t is the time (s); C_V, C_{Vo} and C_{VADJo} are the chemical concentration in the vapor phase of the air compartment, in ambient air and in the adjoining room, respectively (μg chemical/m^3 air); Q_{mv}, Q_{ra}, Q_{cf} and Q_{ADJo} are the volumetric flow rate of air drawn to the mechanical ventilation system, of return air to the mechanical ventilation system, of air through cracks and from the adjoining room, respectively (m^3/s); Q_{window} is the natural air flow through window opening (m^3/s); Q_{rcl} is the air recirculation rate of indoor filter device (m^3/s); e and e_{rcl} are the filter efficiency of the mechanical ventilation system and the indoor filter device, respectively; E_{si} is the rate of emission from interior surface *i* (μg chemical/m^2/s); E_{Vj} is the rate of emission from indoor source *j* other than interior surfaces (μg chemical/s); A_i is the area of interior surface *i* (m^2); k_v is the first order reaction rate constant (1/s).

$$V_c\cdot\frac{d[M(d)\cdot C_p(d)]}{dt} = [Q_{mv}\cdot M_o(d)\cdot C_{p_o}(d) + Q_{ra}\cdot M(d)\cdot C_p(d)]\cdot[1-e(d)] + Q_{cf}\cdot M_o(d)\cdot C_{p_o}(d)\cdot P(d) + Q_{window}\cdot M_o(d)\cdot C_{p_o}(d) + Q_{ADJ_o}\cdot M_{ADJ_o}(d)\cdot C_{p_{ADJ_o}}(d) - (Q_{mv} + Q_{ra} + Q_{cf} + Q_{window} + Q_{ADJ_o})\cdot M(d)\cdot C_p(d) - [\sum_i A_i\cdot V_{d,i}(d)\cdot M(d)\cdot C_p(d) + Q_{rcl}\cdot e_{rcl}(d)\cdot M(d)\cdot C_p(d)] + [\sum_k M_{p_k}(d)\cdot C_{p_k}(d) + v_r(d)\cdot M_f(d)\cdot C_f(d)\cdot A_d] \quad (2)$$

where d is the aerodynamic diameter of PMs (μm); M(d), M_o(d) and M_{ADJo}(d) are the PMs mass concentration in the air compartment, ambient air and the air of adjoining room, respectively (μg PMs/m^3); C_p(d), C_{po}(d) and C_{PADJo}(d) are the chemical mass sorbed to PMs in the air compartment, ambient air and the air of adjoining room, respectively (μg chemical/μg PMs); e(d) and e_{rcl}(d) are the filter efficiency of the mechanical ventilation system of PMs and the indoor recirculation device of PMs, respectively; P(d) is the penetration factor of PMs through cracks; A_d is the total area of the downward-facing interior surface (m^2); $V_{d,i}$(d) is the PMs deposition velocity from indoor air to surface *i* (m/s); V_r(d) is the PMs resuspension rate coefficient from surface to indoor air (1/s); M_{Pk}(d) is the PMs emission rate from indoor source *k* (μg PMs/s); M_f(d) is the PMs concentration on the floor surface

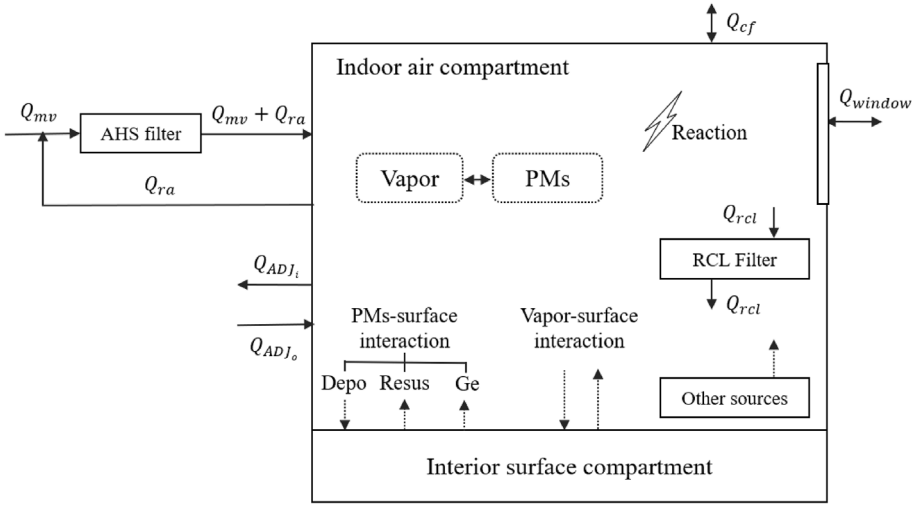


Fig. 1. Schematic representation of IIAQ-CC (Q: volumetric air flow rate, mv: mechanical ventilation, cf: crack flow, ra: return air, RCL: recirculation, ADJ_i: to adjoining room, ADJ_o: from adjoining room, AHS: air handling system, PMs: particulate matters, Depo: deposition, Resus: resuspension, and Ge: generation).

(μg PMs/m^2); C_{Pk}(d) and C_f(d) are the chemical concentration associated with M_{Pk}(d) and M_f(d), respectively (μg chemical/μg PMs).

Equation (2) could be solved either for each PMs size range or for a lumped PMs (e.g., total suspended particles) with no size resolution. For size dependent calculation, the entire PMs range should be defined first and the range is divided to create sub-ranges of equal interval of one log scale. Each sub-range is further divided into 10 equal intervals in log scale. It was assumed that the PMs size distribution, once set, does not change. In the present work, the FA concentration in the PMs phase was calculated to be negligible as the FA's sorption coefficient is substantially small.

2.1.2. Model structure

The target pollutants of IIAQ-CC are VOCs and SVOCs. IIAQ-CC consists of 7 modules and 34 subroutines as shown in Fig. 2. All the parameter and variable types are defined in the Global Variables module. To run a simulation, a number of conditions should be specified in the Initial Conditions module including the CC scenario, simulation period, a pollutant of interest, a modeling location within South Korea, and others such as chemical phase selection (vapor only, PMs only, or vapor-PMs), and process options in accordance with other simulation conditions (e.g., no PMs deposition/resuspension in the "vapor only" mode or PM size range in the modes of "PMs only" and "vapor-PMs"). Once a model location and a CC scenario are selected, IIAQ-CC receives modeled meteorological data and ambient air concentration data for the selected area (Data Input module). More details of these data are described in section 2.2. As a primary model input, a number of parameters (Table A.2) should be directly entered into the

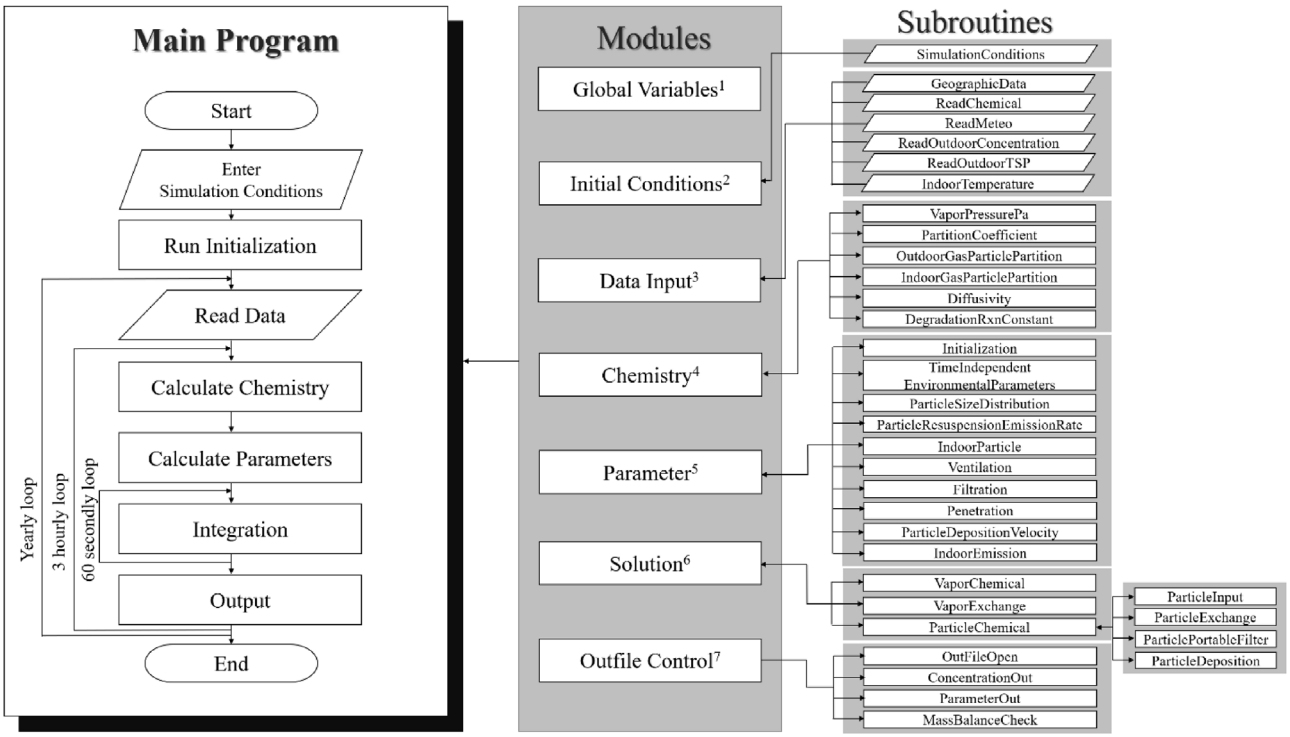


Fig. 2. Structure of IIAQ-CC ([1]defining variable types, [2]specifying initial simulation conditions, [3]reading the input data, [4]calculating chemical characteristic variables, [5]calculating various other variables, [6]solving a set of mass balance equations, [7]printing simulation results).

Table 1
Temperature dependent variables used in IIAQ-CC.

Variable name	Unit	Description	Equation	[a]Related process	Reference
AirViscosity	kg/m/s	Dynamic viscosity of air	$\mu = \rho_a(T)\cdot \nu(T)$	D F P	[28]
[b]DeltaPressure	Pa	Pressure difference	$\Delta P = \left(\frac{1}{2}C_p\rho_{out}U_{ref}^2\right) + \left\{\rho_{in}g(H - H_{npp})\frac{T_{in}-T_{out}}{T_{out}}\right\}$	V P	[29,30]
[c]DiffusionInMaterial	m^2/s	Effective diffusion coefficient of a chemical in the interior surface *i*	$D_i = 10^{-10}\,T^{1.25}\exp(-4109.1/T)$ for formaldehyde in vinyl floor	V-S	[31]
[d]DiffusivityInAir	m^2/s	Diffusivity of a chemical in air	$D_a = 10^{-3}\frac{T^{1.75}[1/MW_{air}+1/MW_m]^{1/2}}{P[\bar{V}_{air}^{1/3}+\bar{V}_m^{1/3}]^2}\cdot(10^{-4})$	V-S	[32]
DischargeCoeff	–	Discharge coefficient for openings	$C_d = (0.4 + 0.0075\lvert T_{in}\text{-}T_{out}\rvert)$	V	[33]
[e]DpBrown	m^2/s	Particle diffusion coefficient	$D_p = kTC_c/(3\pi\mu d_p)$	D F P	[34]
[f]Fnc_AirDensity	kg/m^3	Air density	$\rho_a = pM/(RT)$	D F P V	[28]
Fnc_AirMeanFreePath	m	Mean free path for air	$\lambda = 2\mu/(p\sqrt{(0.001)\cdot 8MW_m/\pi RT})$	D F P	[34]
Fnc_KViscosityAir	m^2/s	Kinematic viscosity of air	$\upsilon = [1.32 + 0.009\cdot(T - 273.15)]/10\cdot(10^{-4})$	D V-S	[28]
[g]IndoorVP/OutdoorVP	Pa	Indoor/Outdoor vapor pressure	VOCs: $\log(P_v\cdot(10^{-5})) = A\text{-}[B/(T\text{-}C)]$ PAHs, PCDDs/DFs, and PCBs: $\log P_v = -A/T + B$ PBDEs: $\ln(P_v/P_{v,\,ref}) = \Delta_{vap}H\cdot(10^3)/R\cdot(1/T_{ref}\text{-}1/T)$	V-P	[35–37]
Kma	–	Equilibrium partition coefficient between vapor and the interior surface *i*	$K_{ma,\,i} = 0.00005\cdot T^{1/2}\exp(4704/T)$ for formaldehyde in vinyl floor	V-S	[38]

[a] D: deposition, F: filtration, V-P: vapor-particulate partitioning in air, P: penetration, V: ventilation, V-S: vapor-surface interaction.
[b] C_p: pressure coefficient, ρ_{out}: density of outdoor air (kg/m^3), ρ_{in}: density of indoor air (kg/m^3), U_{ref}: wind speed at a reference height (m/s), g: gravitational acceleration (m/s^2), H: height above reference plane (m), H_{NPP}: height of neutral pressure plane (m), T_{in}: indoor temperature (K), T_{out}: outdoor temperature (K).
[c] T: absolute temperature (K).
[d] MW_{air}: mean molecular air weight (g/mol), MW_m: molecular weight of a pollutant (g/mol), P: atmospheric pressure (atm), $\bar{V}_{air}$: mean molar gas volume of the air (cm^3/mol), $\bar{V}_m$: mean molar volume of a pollutant (cm^3/mol).
[e] k: Boltzmann constant ($kg.m^2/s^2/K$), C_c: slip correction factor, d_p: particle diameter (10^{-6}μm).
[f] p: absolute pressure (Pa), M: molar mass (kg/mol), R: ideal gas constant (m^3. Pa/mol/K)).
[g] P_v: vapor pressure (Pa), A,B,C = temperature dependence constant of P_v, $P_{v,ref}$: vapor pressure at T_{ref}, $\Delta_{vap}H$: enthalpy of vaporization (kJ/mol), T_{ref}: reference temperature (K).

Data Input module. As a secondary model input (Table A.2), chemical characteristic variables and various other variables are calculated as a function of temperature by the Chemistry and the Parameter modules, respectively. Indoor pollutant concentration is calculated by solving a set of mass balance equations within the Solution module with a computation time step of 1 min.

2.1.3. CC scenario

Influence of CC was accounted for in IIAQ-CC primarily through atmospheric temperature change predicted in accordance with a selected CC scenario, which subsequently accompanies changes in indoor temperature, occupant's behavior concerning window opening coupled with use of HVAC system, and temperature dependent rates of fate and transport processes indoor listed in Table 1. In the present work, RCP8.5 was selected for the case study of FA. The modeling period was divided into four intervals; 1975 to 2005 (Base) as a base and three CC periods of 2011–2040 (F1), 2041 to 2070 (F2), and 2071 to 2100 (F3).

2.1.3.1. Indoor temperature. Indoor temperature was estimated using three empirical linear relationships between indoor and outdoor temperatures over the outdoor temperature range from −13 °C to 33 °C [27] (Indoor temperature in Table A.3). The relationships for winter, spring/autumn, and summer were used at outdoor temperatures below 4 °C, between 4 °C and 21 °C, and above 21 °C, respectively. The seasonal temperature divides (i.e., 4 °C and 21 °C) were determined to be the outdoor temperature of the highest frequency in each season during the years from 1988 to 2017 [39]. However, no such an empirical relationship was available for the outdoor temperature below −13 °C or above 33 °C. Extrapolation using any empirical relationship beyond the conditions from which the relationship is derived is strongly discouraged. Thus, the indoor temperatures at outdoor temperature outside the temperature range (below −13 °C or above 33 °C) were estimated from the linear relationships at −13 °C for winter and at 33 °C for summer as more than 99.5% of the simulation period is at outdoor temperature between −15 °C and 36 °C. Extrapolation projected that the deviation of the estimated indoor temperature would be less than 0.15 °C and 0.1 °C from those at 33 °C (summer) and −13 °C (winter), respectively. Such small temperature deviations hardly make any difference in the fate and transport of FA indoor as far as window opening status remains unchanged (window closed in any case at temperature < −10 °C or > 30 °C). Based on the indoor temperature determined as above, the temperature dependence of the fate and transport processes was calculated from those of the variables listed in Table 1.

2.1.3.2. Occupant's behavior. In the present work, changes in window opening/closing and use of heater/cooler in response to CC were of concern among occupants' behaviors as the former and the latter could substantially influence the air exchange rate and the indoor temperature, respectively.

In Fig. 3, it was assumed that at the outdoor temperature below 10 °C [11,40,41] or above 30 °C [42], a heating (HT) system or a cooling (CL) system would run. At outdoor temperatures between 10 °C and 30 °C, the indoor temperature would be controlled mainly through window opening/closing (free-running (FR) mode). While the HT or CL system was on, window was assumed to be closed. At temperatures between 10 °C and 30 °C, window opening behavior was determined using a relationship between comfort and the indoor temperatures based on the Humphreys adaptive algorithm [43]. According to the algorithm, comfort temperature is first calculated from a linear relationship between comfort and exponentially-weighted running mean outside temperatures [44]. The linear relationships have been empirically determined based on the adaptive thermal comfort theory [41,45]. It was found in a previous study [43] that over 80% of the window status is not changed within the deadband of ± 2 K from the comfort temperature. Therefore, the ± 2 K deadband was taken in the present work within which the indoor environment is considered comfortable and window status is assumed to remain unchanged.

2.1.3.3. Natural ventilation. In the present work, natural ventilation

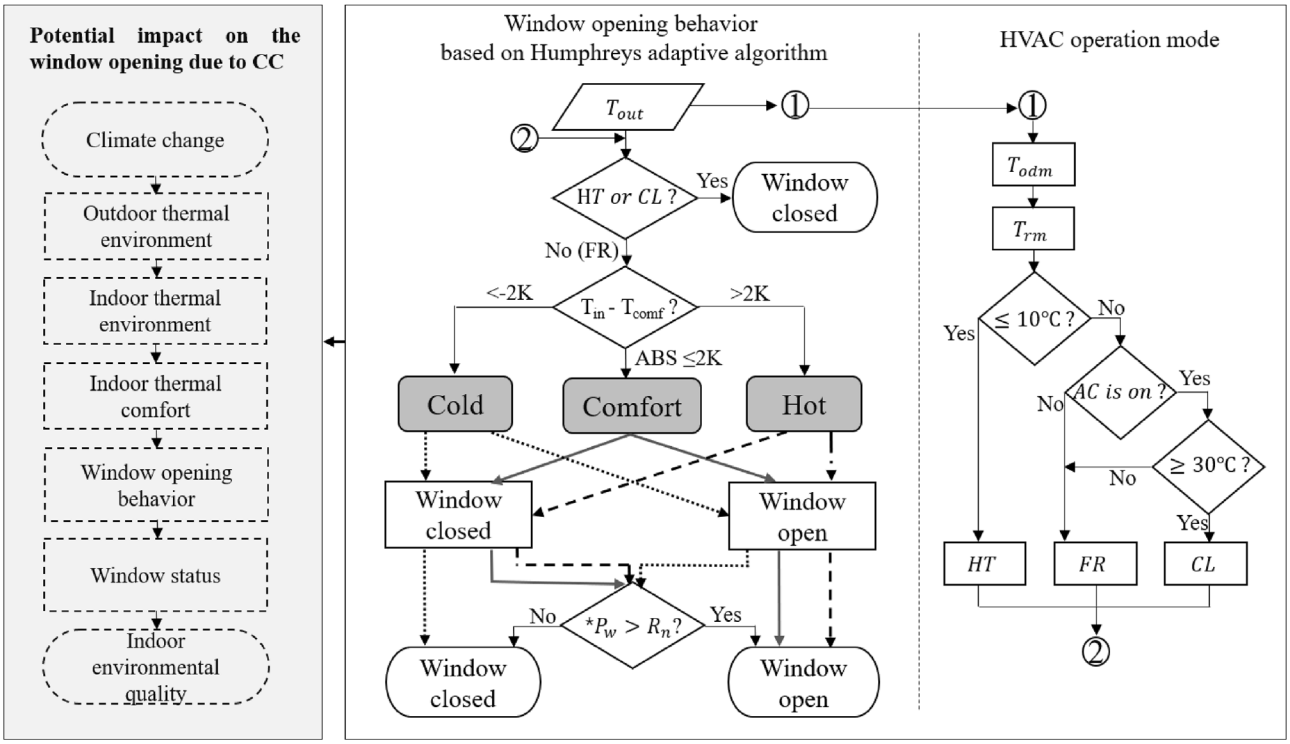


Fig. 3. Sequence in IIAQ-CC to assess CC influences on indoor air quality through window opening behavior (left grey box) and the logics to determine adaptive behaviors (window opening [43] and use of HVAC) (right white box) (CC: climate change, T_{out}: outdoor temperature, T_{in}: indoor temperature, T_{comf}: comfort temperature, T_{odm}: daily mean outdoor temperature, T_{rm}: running mean outdoor temperature, P_w: probability function for window open, R_n: random number generated from a uniform distribution between 0 and 1, HVAC: heating, ventilating, and air conditioning system, HT: heating mode, FR: free-running mode, and CL: cooling mode). *$probability(P_w > R_n) = P_w$.

refers to the sum of air change through window opening and crack. The natural ventilation was calculated using the ventilation equations in Table A.1 and pressure difference in Table 1. It should be noted that natural ventilation rate depends not only on the indoor/outdoor temperature difference but the pressure difference driven by wind [33,46].

2.2. Input data

2.2.1. Meteorological data

Two climate conditions were used; one for the base period of 1956–2005 and the other driven by the CC scenario RCP8.5 for the period from 2011 to 2100. The RCP8.5, adopted by the IPCC in its fifth Assessment Report [1], is a scenario of comparatively high greenhouse gas emission continuing to rise throughout the 21st century [47]. The meteorological data (temperature, wind speed and direction, and precipitation) of South Korea were obtained from the regional downscaling data produced by the National Institute of Meteorological Sciences from the original work done using Global Climate Model [48] (Fig. A1). Temporal and spatial resolutions of the data are 3-h and 30 km × 30 km, respectively.

2.2.2. Outdoor air concentration

Ambient air concentration of FA was predicted with a multimedia fate model, KPOP-CC, which shares meteorological data with IIAQ-CC. The modeling domain of KPOP-CC covers entire South Korea with a spatial resolution of 30 km × 30 km [49]. KPOP-CC has been successfully evaluated against measured data sets of the multimedia environmental levels of various VOCs and SVOCs [49]. The FA concentration in outdoor air of any area of concern in South Korea can be predicted and input to IIAQ-CC.

2.3. FA case study

Before applications of IIAQ-CC for future prediction, it was evaluated for its prediction performance by comparing with indoor monitoring data of FA in air. As FA would well represent most of VOCs of indoor environmental concern, the evaluation results are likely to be applicable to the VOCs. However, further evaluation studies will need for SVOCs because their fate and transport characteristics are substantially different from VOCs', significant interactions with PMs in particular.

2.3.1. Monitoring data

Indoor FA monitoring was conducted to evaluate IIAQ-CC for its prediction performance of CC influence largely driven by temperature change. Preliminary monitoring indicated potentially significant temperature influence on the FA concentration at the outdoor temperature from −15 °C to 20 °C while no clear temperature influence was observed above 26 °C–34 °C. Therefore, a total of 129 gaseous FA samples were collected from 24 households for two periods of December of 2012 to February of 2013 and December of 2014 to February of 2015.

The FA concentration in indoor air of a living room was measured 3 times for each of 18 households and 5 times for each of 15 households in the first and second periods, respectively. Eight of these households were measured for both periods. Air was collected using SKC UMEx100 passive sampler for a 24-h period (OSHA method 1007) [50]. The sampler was installed at 1 m–1.5 m height above the floor and was placed so as to avoid direct influence of any nearby indoor pollution source such as furniture and electronic products. FA was analyzed using a method that partially modifies TO-11A [51] and SKC UMEx100 passive sampler analysis method [52]. The detection limit of FA in air was 0.027 μg/m^3 and the recovery ratio was 93%.

Indoor temperature was simultaneously measured with thermo-hygrometers (Testo 174H) for the 24-h duration of individual sampling.

Outdoor temperature was obtained from the meteorological data of Korea Meteorological Administrator for the sampling periods [53]. The 24-h averages of the indoor and outdoor temperatures were assigned to each corresponding sample for data analysis.

2.3.2. Performance evaluation

Each measured concentration value was a result of combined influences of many factors including not only those in the three categories described in the Introduction section but the age of house in case of FA as its internal emission rate from walls or floor depends on the house age. For evaluating IIAQ-CC, direct point by point comparison against the measured indoor FA concentration values was not possible because IIAQ-CC could not predict the concentration of individual houses as their influential factors were only partially known. Instead, the measured data for a house j were used first to calculate the temperature sensitivity of the house as defined by equation (3)

$$(temperature\ sensitivity)_j = \left[\frac{\partial(C_i/C_{mean} \times 100\,(\%))}{\partial(T_{outdoor,i})}\right]_j \quad (3)$$

where C_i and C_{mean} denote the individual concentration values of each house j (3 or 5 or 8 values) and their mean, respectively. Then model performance was evaluated by comparing the temperature sensitivity predicted using IIAQ-CC with a set of the temperature sensitivity values of the individual houses determined from the measured data. The temperature sensitivity is indicative of how the relative concentration indoor responds to the outdoor temperature. As the temperature sensitivity from the measured data was calculated for each of the houses, within-house variation across sampling in the parameters' influences that are independent of temperature would be maintained minimal, thus their influences might be assumed constant during the multiple samplings (3 or 5 or 8) in each house. In addition, use of the relative concentration could reduce house to house variability of the concentration level owing to normalization effect with respect to the mean value. As atmospheric temperature is a principal driver of CC influences, use of outdoor temperature in the denominator of equation (3) is not only important but consistent with the purpose of performance evaluation of IIAQ-CC.

3. Results and discussion

The lifespan of a typical residential house in South Korea was assumed to be 30 years [54] largely because of considerable profits from housing redevelopment projects, which has facilitated premature demolition of houses over the past 50 years or so. Therefore, simulations were conducted for each of the three 30 year periods of 2011–2040 (F1), 2041 to 2070 (F2), and 2071 to 2100 (F3) and the results were compared with those of the base period of 1976–2005 (Base). The input information used for the simulations is summarized in Table A.4 of the SD.

3.1. Window opening behavior

It is shown in Fig. 4(a) that total duration of window open (DOW, refers to the length of time during which window stays open) for the Base and CC periods is shortest in winter and longer in summer (except in F3) than in spring and autumn. As shown in Fig. 4(b), DOW tends to increase with outdoor temperature. Because outdoor temperature steadily rises as CC progresses from F1 through to F3, increase in DOW of individual seasons should be in the order of F3 > F2 > F1 as compared to Base, which is the case to spring, autumn, and winter. DOW in summer, however, rather decreases with the CC progress because outdoor temperature more often rises high enough to keep a CL system on (e.g., above 30 °C) with windows shut. In spite of CC, DOW only slightly increases in winter. As a combined result of the seasonal DOW changes, the percent change of the annual DOW (= ($DOW_{RCP8.5}$- DOW_{Base})/DOW_{Base} x 100%) in F2 and F3 are statistically significant (ANOVA test, $p < 0.05$) with increments of 4.6% and 3.4%, respectively, while DOW in F1 also appears to increase a little but with no statistical significance. This result indicates that windows will be open slightly longer (< 5%) in the periods under the CC conditions.

3.2. Natural ventilation

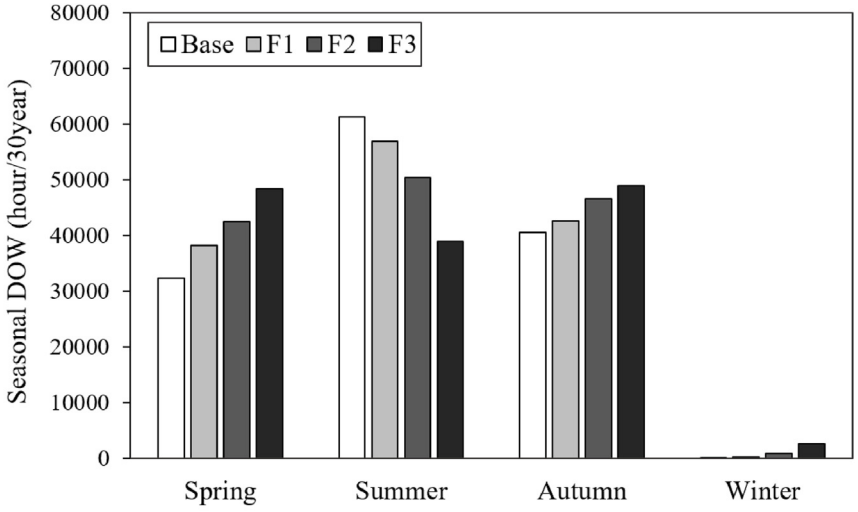
The seasonal average of air change rate (ACR) over each period changes in manners similar to DOW in that it decreases in summer and increases in other seasons as CC progresses because ACR is primarily governed by DOW (Fig. A2). As the result, annual averages of ACR by natural ventilation in F3 are significantly different from those in Base (ANOVA test, $p < 0.05$) with a decrease of 3.0%. The crack flow contributes slightly less than 10% to the natural ventilation (Table A.5). It is noteworthy, however, that while ACR by window opening significantly decreases only in F3, ACR by crack flow steadily declines over the entire period (up to 9.9% in F3) (Table A.5), which is consistent with a previous study [5]. Besides, although the crack flow is only 9.2% of the total natural ventilation in F3, its contribution is 32.3% to the total reduction of ACR (i.e., 3.0%), indicating that the crack flow change should not be neglected for accurately assessing the indoor air quality change caused by CC.

3.3. Model performance evaluation

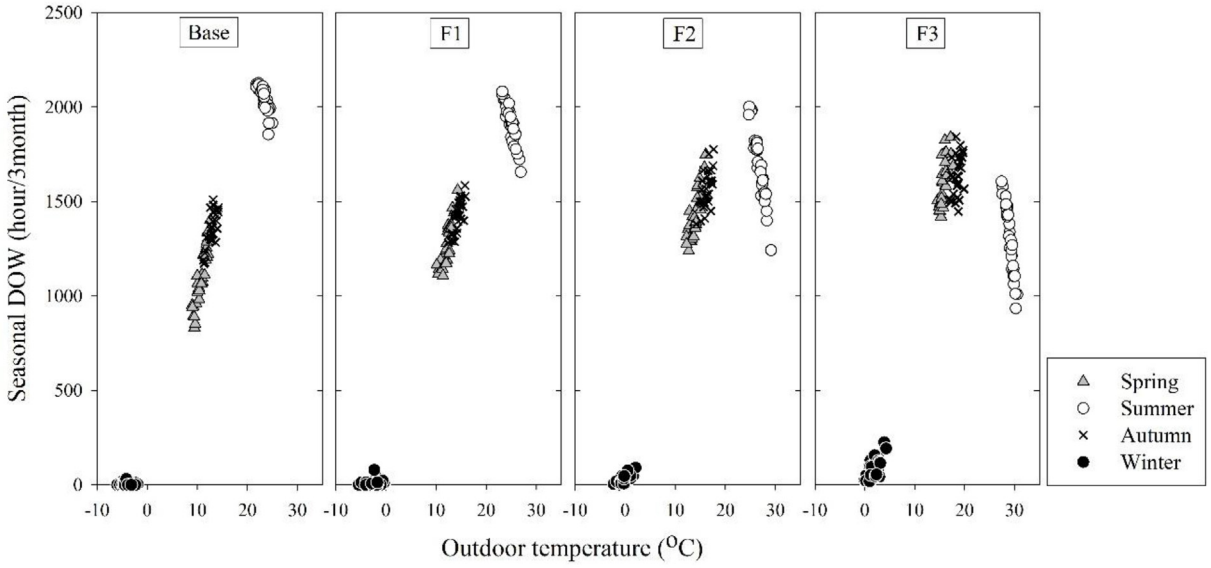
Of the 129 samples collected, 26 samples were found to be contaminated, hence excluded from the data analysis. Also, outliers (> ± 3 standard residual) were removed from the data analysis. As the result, only two data points became available for each of two households, which were further excluded because the temperature sensitivity determined from two data points were not reliable. Finally, a total of 86 measured data from 17 households were used to calculate the temperature sensitivity of individual households (Table A.6).

For model prediction of the temperature sensitivity, the initial concentrations of FA in the air compartment and the vinyl floor (widely used as an indoor flooring material in South Korea) were set to zero and 1.51×10^7 μg/m^3 [38], respectively, to calculate the diffusive emission rate from the floor. The details of characteristics of a living room typical of South Korean houses used for the prediction are summarized in Table A.4 of the SD. A single value of temperature sensitivity was predicted for the living room from the mean of the 24-h averages of indoor air concentrations calculated at every 3 h.

In Fig. 5, all the 86 individual points of C/C_{mean} x100 vs. outdoor temperature are shown for the measured data and compared to the 95% band of the predicted values where both trends indicate that the relative concentration tends to increase with outdoor temperature. The two slope values determined from each of the measured and the prediction data sets are statistically significant (95% confidence intervals of 3.8 ± 0.8%/°C and 3.9 ± 1.0%/°C, respectively). While the slope from prediction is truly the predicted temperature sensitivity of the single living room, that from the measured data set shown in Fig. 5 is not exactly the temperature sensitivity as defined by equation (3). Nonetheless, it was notable that the two slope values are almost identical to each other with no significant difference (t-test, $p < 0.05$), suggesting approximately that the temperature sensitivity predicted by IIAQ-CC will be in good agreement with those calculated from the measured data. In fact, the average of true temperature sensitivities of the individual houses was 5.0%/°C with a 95% confidence interval from 3.7%/°C to 6.4%/°C, which includes the IIAQ-CC predicted value (3.9%/°C). Therefore, IIAQ-CC was assessed to be capable of quantitatively predicting the indoor concentration change of FA in response to outdoor temperature from −15 °C to 20 °C. As the outdoor temperature increases from 20 °C to 26 °C where monitoring data are absent, the temperature dependence was predicted to become gradually dispersed. At outdoor temperature above 26 °C, both prediction and monitoring



(a)



(b)

Fig. 4. Seasonal duration of window open (DOW) for (a) each of the 30 year periods and (b) 3 month periods with respect to outdoor temperature (Base: 1976 to 2005 for base period, and F1: 2011 to 2040, F2: 2041 to 2070, and F3: 2071 to 2100 for RCP8.5 climate change scenario).

data showed no significant temperature dependence although the monitoring data were mostly within the 95% prediction band (Fig. A3).

3.4. Influences of CC on the long-term FA concentration in indoor air

Three indoor emission scenarios were assessed with an assumption of constant atmospheric emission of FA at 0.2 kg/km^2/h throughout the simulation periods: (1) no indoor emission source (ES1), (2) indoor emission only from vinyl flooring (ES2), and (3) periodic indoor emission from a strong pulse source of constant strength (ES3). ES1 assumes that FA in the indoor environment originates only from FA in outdoor air. In ES2, diffusive emission from vinyl floor was added to the ES1 condition. Among a number of occupant activities that have been known as indoor FA sources, cooking fish was selected as an indoor source for ES3 with the ES1 condition as a background. Each fish cooking was assumed to take 20-min with a frequency of twice a day. The emission rate during the cooking was set to 8.35×10^{-2} μg/m^3/s [55], which gave rise to predictions that are within the observed indoor FA range in South Korea (20 μg/m^3 to 60 μg/m^3) [56].

It is shown in Fig. 6(a) that the outdoor FA concentration tends to slowly and steadily increase over the entire simulation period with a statistically significant slope of 0.009 μg/m^3/yr (p = 0.95), indicating that CC in the long run may slightly worsen the ambient air quality (FA) even if the atmospheric emission stays the same. Although the concentration level is lower than that in outdoor, similar time trends appear for the indoor air concentration of FA both under ES1 and ES2 with statistically significant slopes of 0.010 μg/m^3/yr and 0.009 μg/m^3/yr, respectively. With the slopes under ES1 and ES2, the FA concentration increases during F3 by 11.6% and 11.5%, respectively (Table A.7).

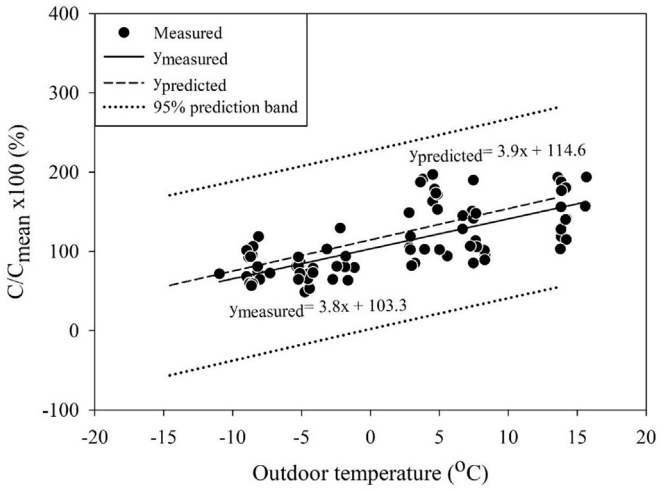


Fig. 5. Predicted temperature sensitivity (with a 95% band) compared to the measured 86 individual values from 17 households.

The time trend under ES1 is comparable to that of outdoor because the FA concentration of indoor is principally governed by that of outdoor. Despite that an additional emission from floor was assumed in ES2, the long-term (up to 30 years) averages of the indoor FA concentration are almost the same as those under ES1 because the emission from floor declines rapidly (e.g., about 2–3 weeks to decay to 10% of the initial rate). In ES1 and ES2 where indoor emission is absent or negligible, therefore, the change in indoor FA concentration is driven by that in outdoor FA concentration. However, the FA concentration level under ES3 is markedly greater than those under ES1 and ES2. Moreover, it is notable that the time trend is opposite with a negative slope of $-0.013\,\mu g/m^3/yr$ (significant at $p = 95\%$) for the FA concentration to decrease by 3.2% in F3. Because no mechanical ventilation was assumed, the FA concentration indoor is primarily controlled by indoor FA emission rate and FA exchange flux through natural ventilation. Among other temperature dependent processes, only photodegradation is not negligible as compared to ACR (Table A.8). The higher FA concentration under ES3 is evidently due to the substantially greater indoor emission rate ($8.35 \times 10^{-2}\,\mu g/m^3/s$ indoor vs. $5.6 \times 10^{-4}\,\mu g/m^3/s$ outdoor). The decreasing trend is because of FA loss over time via natural ventilation.

It is shown in Fig. 6(b) that the FA concentration (under ES3) is substantially greater in winter than other seasons while lowest in summer. The FA concentration is in the reversed order of seasonal DOW, showing that the FA concentration is strongly governed by DOW (or ACR) (Fig. 7). Simulations under ES3 further predict that monthly mean indoor concentration increases as much as 320% (in July of F3) as

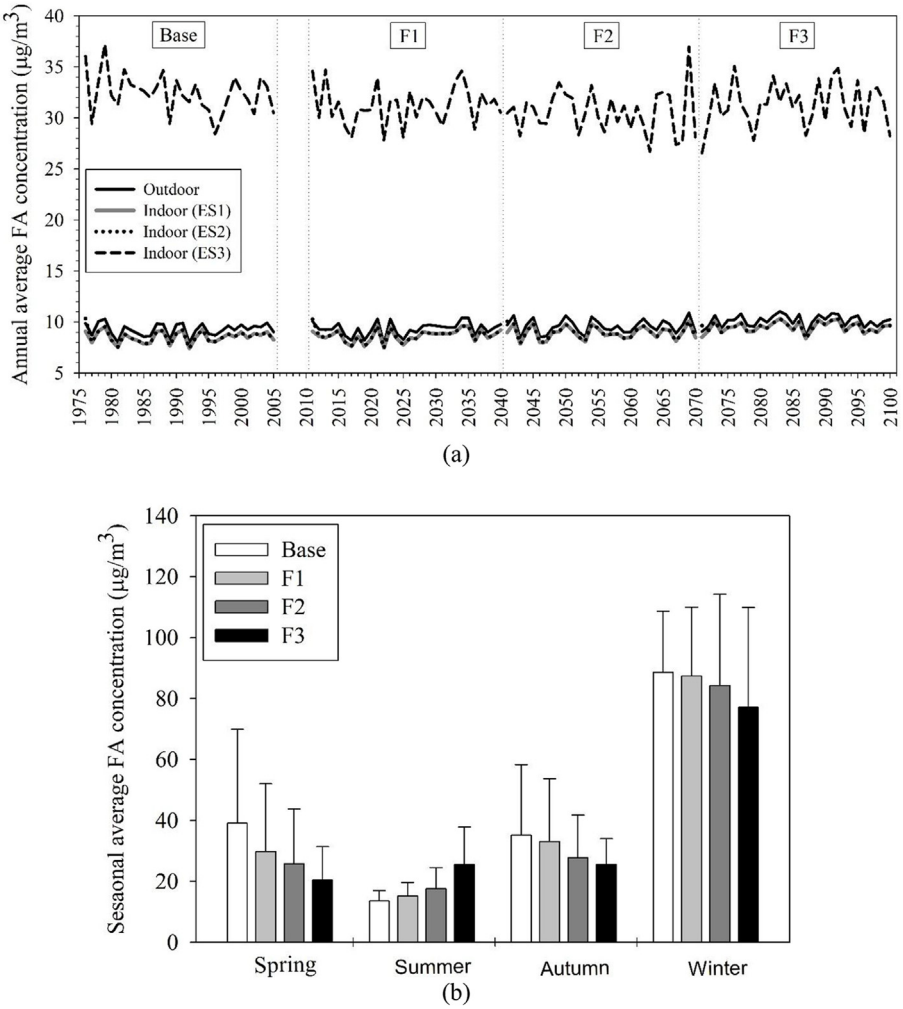


Fig. 6. Effects of CC on the FA concentration; (a) time trend of the annual FA concentration under different indoor emission scenarios (ES) (FA: formaldehyde, CC: climate change, Base: base period, ES1: FA only from outdoor air with no indoor source, ES2: diffusive emission from vinyl floor plus the ES1 condition, and ES3: a periodic strong pulse indoor source plus the E1 condition, Base: 1976 to 2005, F1: 2011 to 2040, F2: 2041 to 2070, F3: 2071 to 2100 for CC (RCP8.5)) and (b) seasonal average of monthly averages over 30 years of FA concentration under ES3 (error bar: one standard deviation).

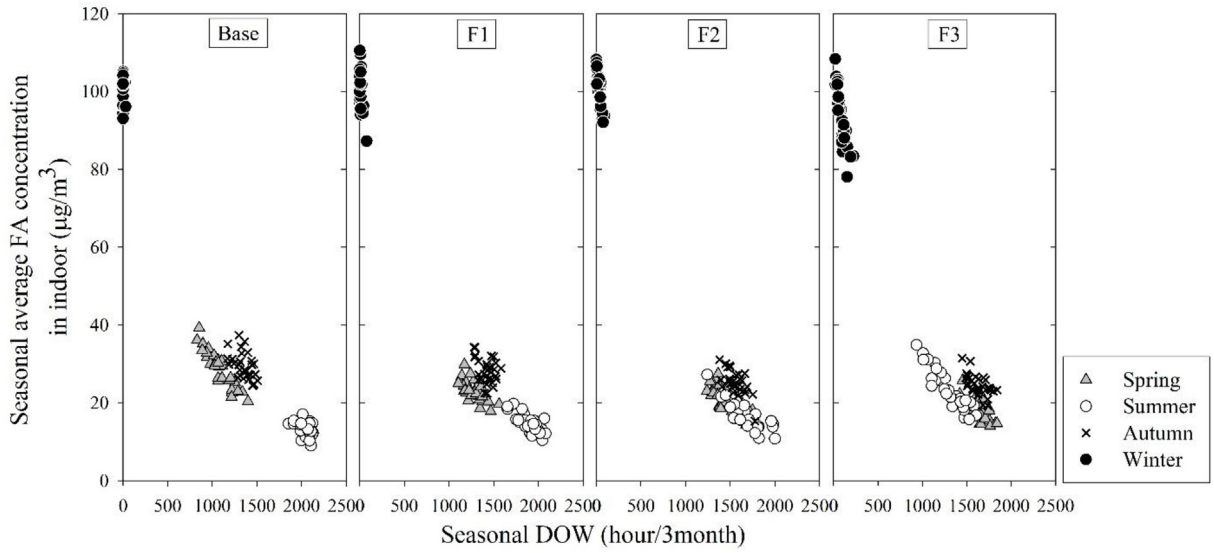


Fig. 7. Seasonal average indoor FA concentration as influenced by seasonal average of DOW under ES3 (FA: formaldehyde, DOW: duration of open window, Base: 1976 to 2005 for base period, F1: 2011 to 2040, F2: 2041 to 2070, F3: 2071 to 2100 for RCP8.5 climate change scenario, and ES3: indoor emission scenario of a periodic strong pulse emission (8.35×10^{-2} μg/m^3/s for 20 min, twice per day)).

compared to that of Base. It suggests that the indoor air quality may substantially degrade during a short period, summer in particular, when windows are kept closed during usage of a CL system.

4. Conclusion

IIAQ-CC was successfully developed to be capable of quantitatively predicting indoor air quality changes (concerning FA and potentially VOCs) with temperature change due to CC. According to the prediction of IIAQ-CC, the long-term FA concentration in indoor air may slowly deteriorate due to the increase in the outdoor concentration (ES1 and ES2) or may decrease in the presence of strong indoor sources (ES3) under the influence of CC (RCP8.5) in South Korea. In either case, however, the long-term (30 years) changes in the indoor concentration of FA do not exceed 12% of the average FA concentration of Base. The FA level may, however, noticeably rise for a short period of time (e.g., month) when windows are closed due to exceedingly hot or cold weather under CC conditions. The FA concentration change, be it significant or not, is primarily governed by DOW, which depends on outdoor temperature. That is, temperature manifests its effects on the indoor air quality essentially by changing adaptive occupant's window opening behavior.

Limitations of the present work that need to be addressed include i) model evaluation against indoor SVOCs such as PBDEs and PAHs, ii) temperature effects through occupants' behavioral change on indoor PMs generation and the consequential PMs level change in indoor air, iii) effect of rain on window opening behavior, and iv) more sophisticated indoor air chemistry when appropriate. All the limitations warrant further studies to improve the model's prediction capability of CC influences.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.buildenv.2018.07.009.

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